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Study of the corrosion behavior of tantalum coatings on AISI316 substrate for bipolar plates of PEM fuel cells

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1. Introduction

Bipolar plates (BPs) are important mechanical elements of PEM fuel cell stacks. They are responsible for both separating and electrically connecting adjacent cells, ensuring a correct distribution of the reactant gases over the catalyst layers, facilitating the water management, and removing the heat excess when dedicated cooling plates are not included. To fulfill all these tasks, both a suitable flowfield geometry design and a correct choice of materials are needed. Materials used in BPs must have high electrical conductivity, low gas permeability, high corrosion resistance, high mechanical strength and low thermal resistance. Satisfying all these conditions can cause an increase in the final cost of the device [1]. Recent cost estimations by the DOE indicate that BPs represent around 25% to 28% of the total cost of stacks, considering a production rate from 100,000 to 500,000 systems per year [2]. Metallic plates and graphite-based composite sheets are the most typical materials used in BPs [3]. The main disadvantage of metallic plates lies in its chemical instability under the PEMFC operating conditions in both anode and cathode sides [4]. To minimize corrosion effects, a protective surface coating has to be deposited, which has to be electrically conductive. Metal nitrides and carbon-based coatings have been reported to meet both corrosion current density and contact resistance DOE targets by ex-situ experiments [5]. In the present research, a substrate of AISI316 stainless steel coated by a 30 µm tantalum layer, deposited by Chemical Vapor Deposition (CVD) has been tested as an alternative to graphite plates. To this end, the corrosion behavior of this coating has been studied by combining corrosion tests with roughness anlysis, as well as interfacial contact resistance (ICR) and contact angle measurements performed before and after the corrosion tests.

2. Experimental

2.1 Preparation of the samples

Cylindrical probes of AISI316 stainless steel with a diameter of 15 mm and 4 mm thick were manufactured. A tantalum coating (Tantaline - Denmark) of 30 µm minimum thickness was applied to the polished samples by CVD.

2.2 Corrosion tests

Corrosion tests were performed with a three-electrode corrosion cell. An Ag/AgCl electrode was used as the reference electrode, a Pt electrode as the counter electrode, and Ta CVD coated samples as the working electrode. To simulate the anodic conditions, a solution of H_2SO_4 + 0.1 ppm HF (pH 3) at 80°C saturated with N_2 was used as the electrolyte. A potentiodynamic polarization (PP) between -0.4 V and + 0.6 V (vs Ag/AgCl) at 0.1 mV/s was applied to the probes. On the other hand, to simulate the cathodic conditions, samples were exposed to a polarization of +0.6 V during 8 h, using the same electrolyte, but saturating it with air. Experiments were carried out with a PARSTAT 2273 potentiostat-galvanostat, controlled with the PowerSuite software.

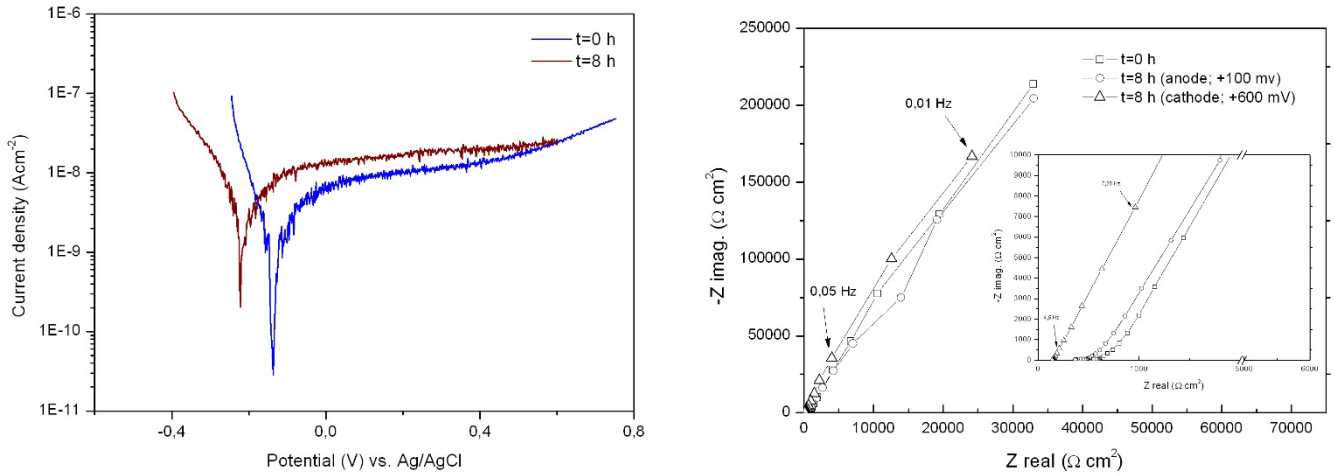
2.3 ICR, contact angle and surface roughness measurements

Before and after potentiodynamic tests, measurements of ICR, contact angle and surface roughness were performed. In addition, as a reference, a sample of polished AISI-316 stainless steel (Ra = 0.2 µm) and a sample of graphite were also tested. ICR measurements were accomplished by placing the samples in sandwich-type configurations, between two layers of carbon paper, which performed the functions of GDL. A manual test bench (AST-J form TECNIMETAL) was used to ensure the correct support-sample assembly during the tests. Contact angle was measured using an optical contact angle OCA 20 device with a droplet volume of 5 µL of deionized water at 20 °C. Surface roughness of the samples was measured with a Mitutoyo Surftest SJ-301.

3. Results and Discussion

3.1 Corrosion tests

Fig. 1.a shows the potentiodynamic polarization curves of two Ta coated samples under anodic conditions for two different times. Both samples show corrosion current densities, I_{corr}, well below DOE targets (<1x10^{-6} Acm^{-2}), with absence of active-passive transition zone. On the other hand, the results of the EIS tests obtained for samples underwent to a similar treatment (Fig. 1b), show a surface with high polarization resistance, thus supporting the results obtained in the DC tests.



a) b)

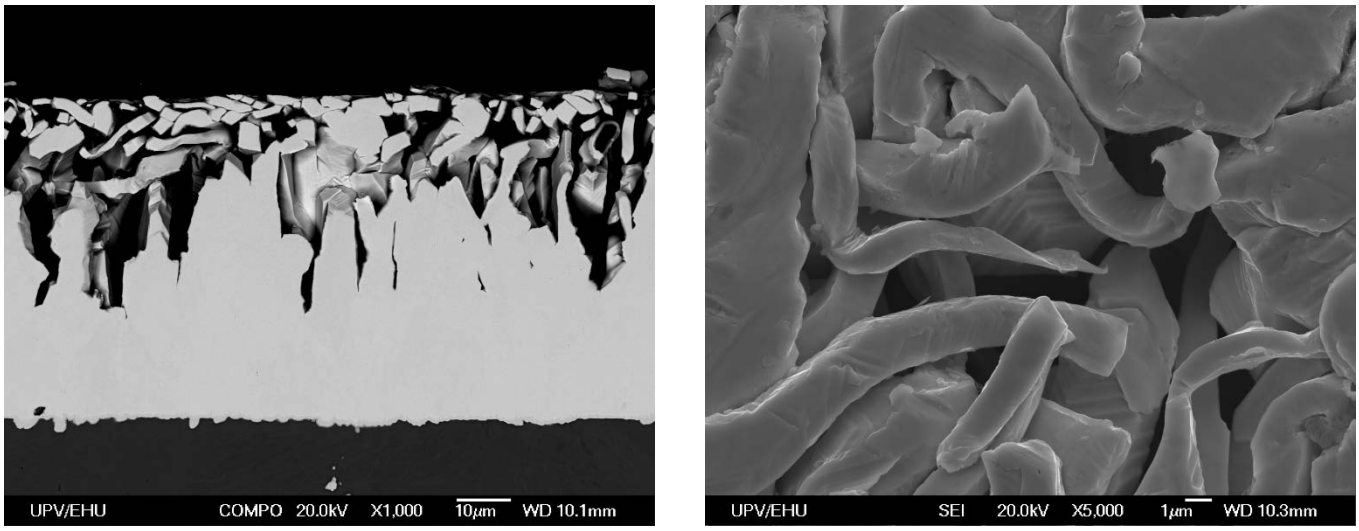
Figure 1. a) Potentiodynamic polarization curves of Ta coatings for anodic conditions. b) Nyquist plots of EIS data obtained from Ta coated samples at different time and treatments of exposure to electrolytic solution (anode: H_2SO_4 + 0.1 ppm HF (pH 3) at 80°C, deaerated with N_2 purge; cathode: H_2SO_4 + 0.1 ppm HF (pH 3) at 80°C, deaerated with air purge).

3.2 ICR, contact angle and surface roughness

The ICR values of the different samples always decrease as the compaction pressure increases, due to an increase of the microscopic contact area. Moreover, comparing the experimental values of ICR after the corrosion tests, it was verified that the ICR does not vary significantly. However, the tantaline layer causes a slight increase in ICR. On the other hand, the surface roughness of the coatings (Ra = 0.4-1.2 µm) is not affected by the corrosion tests, suggesting that there are no major changes of the surface morphology due to the corrosion process. The same behaviour was also observed for the contact angle, which was always above 90º.

3.3 SEM-EDX

Fig. 2 shows examples of both cross-sectional and surface SEM images of the Ta coatings. Results confirmed the absence of superficial morphological modifications after the corrosion tests on the Ta coating layer thickness (30 µm). In addition, the increase of ICR values of the Ta-coated probes could be probably attributed to the columnar morphology observed in these samples. The existence of Ta surface oxides, after corrosion tests, was confirmed by EDX analysis.



a) b)

Figure 2. Cross-sectional (a) and coating surface (b) SEM images of the Ta coatings.

4. Conclusions

The results obtained from both, EIS and PP measurements, show that the Ta coating improves the protection against surface corrosion of AISI-316 stainless steel. Besides, SEM images showed no significant changes in coating morphology after the polarization corrosion tests. Formation of a protective Ta oxide passive film was verified by EDX surface characterization. The combination of high contact angle values (high hydrophobic character [6]) and low ICR values after the corrosion tests, indicate that an AISI316 substrate with Ta coating can be suitable to be used in BPs for PEMFCs.

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